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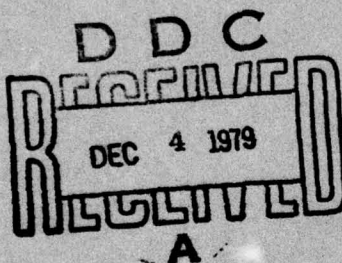
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Thermal Initiation of High Explosives with an Electron Beam

A. STOLOGY, E. C. JONES, JR., J. B. AVILES, JR., AND A. I. NAMENSON

*Radiation-Matter Interactions Branch
Radiation Technology Division*

November 8, 1979



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20. ABSTRACT (Continue on reverse side if necessary and identify by block number) The conditions under which high explosives undergo thermal initiation were studied under dynamic conditions by measuring the temperature of samples while they were irradiated by 40-MeV electrons at the NRL Linac. Values were obtained for the thermal initiation threshold and the explosion temperature for the following high explosives: Composition B, TATB, HBX-1, PBX-9404 and Pentolite. Theoretical values based upon a single rate Arrhenius formula are in reasonable agreement with the experimental results.		

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CONTENTS

INTRODUCTION	1
EXPERIMENTAL PROCEDURE	1
EXPERIMENTAL DATA	3
RESULTS AND DISCUSSION	8
CONCLUDING REMARKS	10
REFERENCES	10

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THERMAL INITIATION OF HIGH EXPLOSIVES WITH AN ELECTRON BEAM

INTRODUCTION

The interaction of a high-energy electron beam with a high-explosive (HE) target offers a new technique for studying the thermal initiation process under dynamic conditions. In more conventional methods, an explosive is placed in a thermal bath or oven so that thermal conductivity plays a complicating role, and the results depend upon the size and shape of the sample. [1,2]. Since a high-energy electron beam is very penetrating, a small sample is heated uniformly throughout its volume, and we can observe the intrinsic thermal behavior of the material. Information on the thermal behavior of explosives is found in some handbooks [3,4], and general properties of explosives are given in other handbooks [5,6].

We are performing a series of experiments in which confined and unconfined samples of high explosives of military interest are irradiated by a 40-MeV electron beam. Our purpose is to obtain the conditions under which a thermal initiation will occur; i.e., how much deposited energy is required to start a runaway exothermic reaction, and at what temperature this will occur. Data have been obtained for five high explosives: Composition B, TATB, HBX-1, PBX-9404, and Pentolite.

EXPERIMENTAL PROCEDURE

The data were taken at the NRL 60-MeV Linac, which produces a pulsed beam of monoenergetic collimated electrons, with pulse repetition rate up to 360/s, and pulse width up to 1 μ s, which delivers about 15 J/pulse. This deposits energy in an explosive sample at the rate of about 130 cal/g-s. The samples are in the shape of disks, 4.3 mm in diameter and weighing between 50 mg and 120 mg. They are contained in an aluminum cup, 0.25-mm-wall thickness, with a plug and clamp to provide confinement, as shown in Fig. 1. A fine-wire (0.001-in. diam) iron-constantan thermocouple is imbedded into each sample. In some cases, the thermocouple is sandwiched between two disks of HE pressed powder. The thermocouple leads are brought out through tiny holes drilled in the HE and plug, and sealed with epoxy. The samples and aluminum cups were obtained from the Naval Surface Weapons Center.* A computer-based data acquisition system is used to obtain temperature readings between beam pulses (every 2.78 ms), so that the temperature can be observed as a function of time during the irradiation until explosion occurs. The response time of the thermocouples is comparable to the time between pulses. Some data were also taken with unconfined samples, where there is no explosion since gases from the decomposing explosive can vent. Nevertheless, many interesting properties of the explosive can be observed, such as phase changes and exothermic reactions.

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*We are indebted to P. J. DiBona and J. W. Forbes for providing us with these materials.

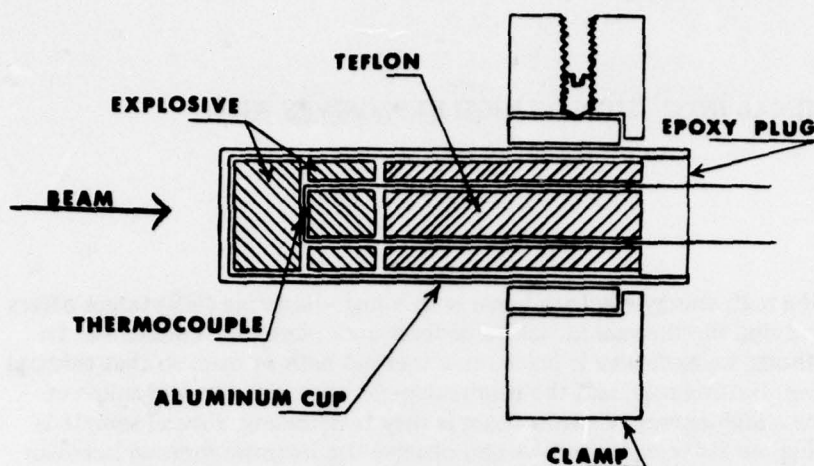


Fig. 1 — Aluminum cup loaded with HE sample and thermocouple

The total dose absorbed by the sample is obtained by calorimetry as follows: an aluminum disk approximately the same size as the sample is placed immediately in front of the sample. The beam passes through this disk with negligible absorption. A small thermocouple is swaged into the aluminum disk, so that its temperature can be observed as a function of time along with that of the sample. The response of an aluminum calorimeter to the beam is shown in Fig. 2. The temperature rises linearly with time until the beam is turned off, whereupon it cools slowly. Since the heat capacity of aluminum as a function of temperature is well known, the beam heating rate can be obtained quite accurately. To apply this to the sample, we make small corrections for beam divergence and for the difference in stopping powers ($1/\rho \cdot dE/dx$) between aluminum and a HE. We also obtain the beam heating rate directly from the explosive, although its heat capacity as a function of temperature usually is not as well known. Sometimes, the data from the explosive are obviously unreliable because of premature separation of the thermocouple from the explosive, resulting in poor contact. Such data were discarded.

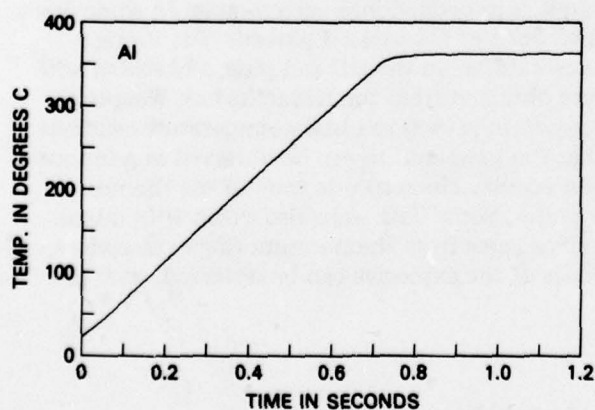


Fig. 2 — Temperature response of aluminum calorimeter to electron beam at 360 pulses/s

By multiplying the beam heating rate by the irradiation time until explosion occurs, we obtain the total deposited dose required for explosion; i.e., the thermal initiation threshold. We also obtain the temperature at which explosion occurs. These results can be compared with values obtained theoretically from the kinetic parameters of the explosive, when these are known.

EXPERIMENTAL DATA

Data obtained from both the HE and the aluminum calorimeter are shown in the following tables. The blanks indicate cases where the data were unreliable due to poor contact between the thermocouple and explosive. The experimental uncertainty in the heating rates for the HE is about 10%, primarily due to uncertainty in the heat capacities. The uncertainty in the heating rates for the calorimeter and the times until explosion is about 1%. The following is a brief discussion of the data for each explosive:

Composition B: In Fig. 3, we observe the temperature response of a confined sample of Composition B to the Linac beam. This explosive is a mixture of RDX, TNT, and wax. The melting of the TNT component at about 81°C is observed as a plateau. When explosion occurs, the thermocouple leads are broken, so no data were obtained in the exothermic region. The total time until explosion is about 1/2 s at a pulse rate of 360 pulses/s. The results of tests on nine samples of this material are listed in Table 1.

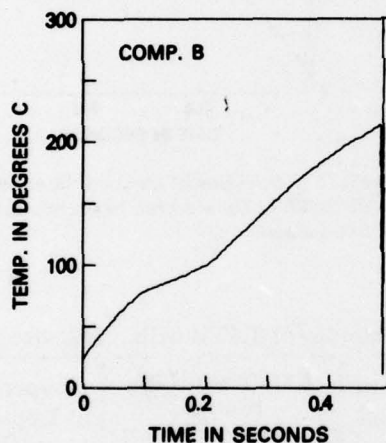


Fig. 3 — Behavior of a confined sample of Composition B under electron beam irradiation (360 pulses/s)

Table 1 — Thermal Initiation of Confined Samples of Composition B with an Electron Beam.*

Heating rate (Cal/g-s)		Time until Explosion (s)	Explosion dose (cal/g)		Temperature at Explosion (°C)
HE	Calorimeter		HE	Calorimeter	
132	115	0.53	70	61	255
—	104	0.53	—	56	—
125	109	0.55	69	60	231
—	113	0.55	—	62	—
—	118	0.54	—	64	—
136	131	0.51	70	67	224
164	125	0.47	77	59	291
192	129	0.48	92	62	270
182	129	0.48	88	62	215

*The pulse repetition rate was 360/s in all cases.

TATB: This material is known to be more difficult to ignite than most military high explosives. However, it undergoes thermal initiation readily with the deposition of somewhat more energy, as shown in Fig. 4 for a confined sample. The response of an unconfined sample of TATB is shown in Fig. 5. There is no violent explosion since the decomposition gases can vent. However, exothermic reactions can be seen as rapid increases in slope, after which a plateau appears indicating melting of this material at 455°C. After melting, further exothermic reactions occur. A summary of results on nine confined samples of TATB are listed in Table 2.

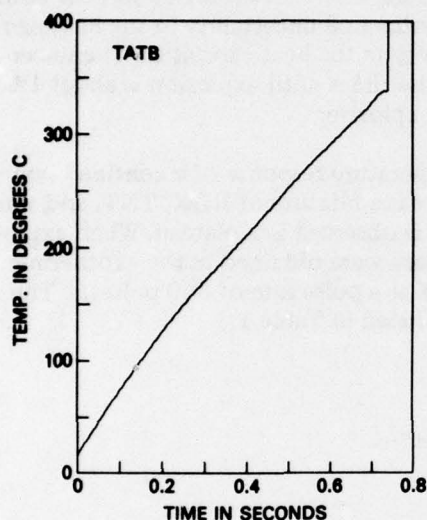


Fig. 4 — Behavior of a confined sample of TATB when irradiated by the Linac beam at 360 pulses/s

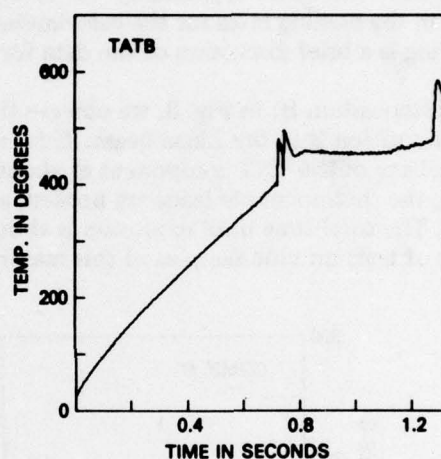


Fig. 5 — Behavior of an unconfined sample of TATB under electron beam irradiation at 360 pulses/s

Table 2 — Thermal Initiation of Confined Samples of TATB with an Electron Beam

Rep rate (s) ⁻¹	Heating rate (Cal/g-s)		Time until Explosion (s)	Explosion Dose (Cal/g)		Temperature at Explosion (°C)
	HE	Calorimeter		HE	Calorimeter	
360	119	131	1.14	136	149	406
360	119	118	0.76	90	90	—
360	113	107	0.75	84	80	334
360	—	120	0.73	—	88	—
360	—	114	0.93	—	106	—
360	100	109	0.77	75	82	315
360	—	108	0.76	—	82	—
180	—	62	1.27	—	78	—
60	23	23	3.53	79	80	313

HBX-1: This mixture is very similar to Composition B, but it also contains aluminum powder. Its behavior, under electron beam irradiation, is almost the same as for Composition B, which shows melting of the TNT component as seen in Fig. 6 for a confined sample. The results of experiments with nine samples of this material are listed in Table 3.

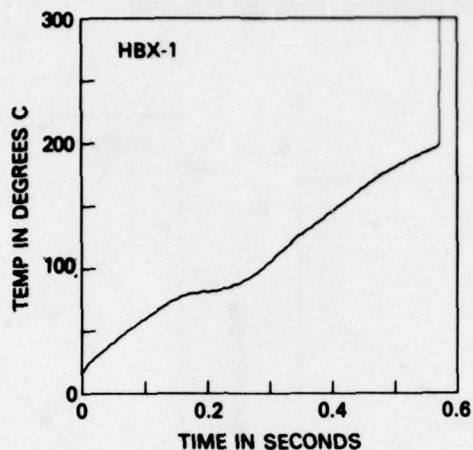


Fig. 6 — Behavior of a confined sample of HBX-1 under electron beam irradiation at 360 pulses/s

Table 3 — Thermal Initiation of Confined Samples of HBX-1 with an Electron Beam

Rep rate (s) ⁻¹	Heating rate (cal/gm-s)		Time until Explosion (s)	Explosion Dose (Cal/g)		Temperature at Explosion (°C)
	HE	Calorimeter		HE	Calorimeter	
360	121	119	0.50	60	59	236
360	114	107	0.60	69	65	195
360	120	103	0.58	69	60	195
360	156	106	0.53	83	56	240
360	135	126	0.57	77	72	237
180	78	67	0.98	76	66	273
60	21	22	2.97	61	66	206
60	18	21	4.10	75	87	250
60	23	21	3.30	75	71	238

PBX-9404: This is a plastic-bonded explosive consisting mostly of HMX and a little nitrocellulose. In the data for a confined sample shown in Fig. 7, we can see some evidence for reaction of the nitrocellulose component near 200°C and melting of the HMX component and explosion at about 275°C. The behavior of an unconfined sample is shown in Fig. 8, where an exotherm occurs after the melt at about 275°C, followed by another phase change plateau at about 400°C, where the liquid HMX vaporizes. The results for four confined samples are listed in Table 4. These are earlier data for which we did not use a calorimeter, so the data are obtained from the HE only.

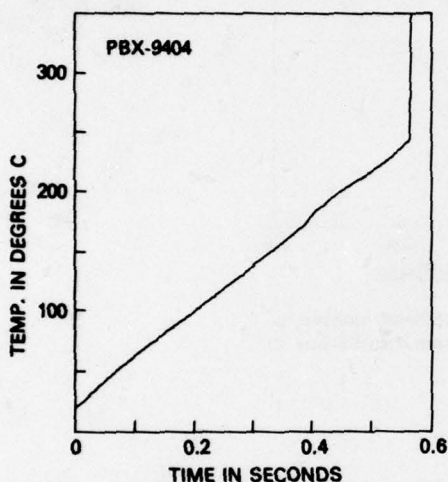


Fig. 7 — Behavior of a confined sample of PBX-9404 under electron beam irradiation at 360 pulses/s

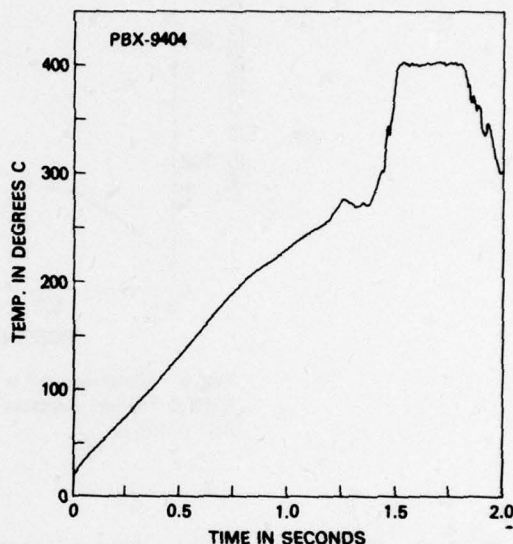


Fig. 8 — Behavior of an unconfined sample of PBX-9404 under electron beam irradiation at 360 pulses/s

Table 4 — Thermal Initiation of Confined PBX-9404 Samples with an Electron Beam

Heating rate (cal/g-s)	Time until Explosion (s)	Explosion Dose (cal/g)	Temperature at Explosion (°C)
16	4.25	68	289
22	2.72	59	252
104	0.61	63	265
117	0.57	67	245

Pentolite: This is a mixture of PETN and TNT which can be initiated more easily than the other explosives we have studied in this report; it usually produces a flare. An example of the response of a confined sample is shown in Fig. 9, where we can see melting of the TNT component, and explosion at a rather low temperature. The data on eight confined samples are listed in Table 5. We have eliminated all data for which the ratio of calorimeter to HE heating rates was greater than 1.5, since this indicates poor contact between the thermocouple and HE.

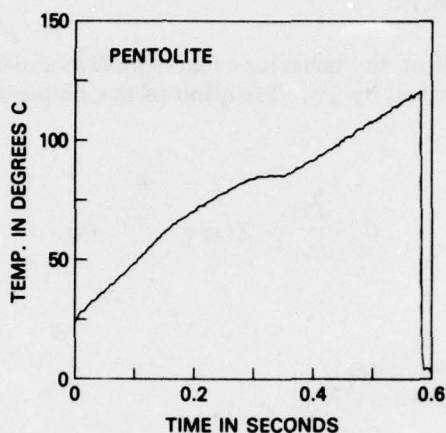


Fig. 9 — Behavior of a confined sample of Pentolite under electron beam irradiation at 360 pulses/s

Table 5 — Thermal Initiation of Confined Samples of Pentolite with an Electron Beam*

Heating rate (cal/g-s)		Time until Explosion (s)	Explosion dose (cal/g)		Temperature at Explosion (°C)
HE	Calorimeter		HE	Calorimeter	
60	83	0.58	35	48	—
—	51	0.94	—	48	—
—	71	0.99	—	70	—
—	71	0.85	—	60	—
—	96	0.68	—	65	—
91	103	0.52	47	53	200
60	80	0.57	34	45	138
85	86	0.51	44	43	178

*The pulse repetition rate was 360/s in all cases.

Data were taken at different beam heating rates in the cases of TATB, HBX-1, and PBX-9404. An examination of the data in Tables 2, 3, and 4 indicates that the explosion dose and the temperature at explosion are essentially independent of the beam heating rate. In the cases of TATB and HBX-1, data were taken with two different sample masses (60 mg and 120 mg). The results indicate no dependence of the thermal explosion threshold or explosion temperature on the mass of the sample. In all cases, the sample cross-sectional area was smaller than the area covered by the beam.

RESULTS AND DISCUSSION

To a first approximation, the behavior of an explosive under uniform irradiation by an electron beam can be described by a modification of the simple Arrhenius equation [1,2] as follows:

$$C_v \frac{dT}{dt} = ZQwe^{-\frac{E_A}{RT}} + \dot{q}, \quad (1)$$

where

- C_v = heat capacity (cal/g-deg),
- T = absolute temperature ($^{\circ}$ K),
- t = time (s),
- Z = collision frequency (s^{-1}),
- Q = heat of reaction (cal/g),
- w = mass concentration of unreacted explosive,
- E_A = activation energy (cal/mole),
- R = gas constant (1.987 cal/mole-deg),
- \dot{q} = beam heating rate (cal/g-s).

We have assumed that thermal gradients are negligible since the beam heats the sample almost uniformly. From this, it follows that the temperature at which the chemical energy heating rate is equal to the beam heating rate is

$$T_e = \frac{E_A/R}{\ln(ZQ/\dot{q})}. \quad (2)$$

This is approximately the temperature at which explosion occurs. The explosion dose (or thermal explosion threshold) is then given by

$$q_e = \bar{C}_v (T_e - T_0), \quad (3)$$

where T_0 is the initial temperature. This represents an upper limit on the energy required to bring about an explosion.

In Table 6, we have listed the kinetic parameters and average beam heating rates which we have used to calculate explosion temperatures and thermal initiation thresholds from Eqs. (2) and (3). The kinetic parameters E_A , Z , Q , and ZQ were obtained from Ref. 4 (pp 4-4, 10-2, and 12-12), Ref. 6, and E. D. Loughran [7] for TATB. The uncertainties of these values are not given. Not all of the required parameters were available, so it was necessary to improvise in many cases. Thus, for HBX-1, we have used the parameters for Composition B, since these explosives are similar. In the case of PBX-9404, we have used the parameters for HMX which constitutes 94% of this explosive. For Pentolite, we have used the parameters for PETN, which constitutes half of this mixture, and is easier to initiate than the TNT component. Average beam heating rates, \bar{q} , were obtained from Tables 1 through 5.

Table 6 — Kinetic Parameters and Average Beam Heating Rates used to Calculate Explosion Temperatures and Thermal Initiation Thresholds.

HE	E_A (kcal/mole)	Z (s ⁻¹)	Q (kcal/g)	ZQ (kcal/g-s)	\bar{q} (cal/g-s)
Comp. B.	44	—	—	6×10^{18}	134
TATB	59.9	3.2×10^{19}	1.20	—	98
HBX-1	44	—	—	6×10^{18}	82
PBX-9404	52.7	5.0×10^{19}	1.48	—	65
Pentolite	47.0	6.3×10^{19}	1.49	—	78

Experimental average values for thermal explosion thresholds and explosion temperatures are compared to the calculated values in Table 7. The experimental average values are obtained by averaging the results obtained from the calorimeter and from the explosive itself. The quoted uncertainties indicate the spread between these two results, or the standard deviation, whichever is larger. The heat capacities of most high explosives are not well known in the higher temperature regions. We have used an average value of 0.27 cal/g-°C for all cases in calculating thermal initiation thresholds from Eq. (3).

Table 7 — Comparison between Experimental and Calculated Values for the Temperature at Explosion and Thermal Initiation Threshold

HE	Thermal Initiation Threshold (cal/g)		Temperature at Explosion (°C)	
	Experiment	Calculation	Experiment	Calculation
Comp. B	69 ± 9	76	248 ± 16	306
TATB	93 ± 6	92	342 ± 22	365
HBX-1*	69 ± 4	75	230 ± 15	299
PBX-9404**	65 ± 9	68	263 ± 15	276
Pentolite***	47 ± 9	52	172 ± 20	216

*Calculations done using kinetic parameters for Comp. B.

**Calculations done using kinetic parameters for HMX.

***Calculations done using kinetic parameters for PETN.

CONCLUDING REMARKS

The theory of thermal initiation which we have used here is of a rather elementary form. A single rate Arrhenius formula may not be a good description of the exothermic region, especially for mixtures. Even for a single type high explosive, there may be several reactions going on, each at a different rate. Nevertheless, as far as explosion temperatures and thermal initiation thresholds are concerned, the theory should yield reasonable values. Indeed, agreement between the experimental and calculated values in Table 7 is good in most cases. The most serious disagreements are for the temperature at explosion for Composition B and HBX-1, for which the calculations were done using the same kinetic parameters (listed in Table 6). This indicates that these kinetic parameters may be incorrect.

We are now performing experiments in which both temperature and gas pressure in the exothermic region can be studied in detail. This will make it possible to evolve a theory which would fit the data more accurately.

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